PREFACE

Quantum-coherent energy transfer: implications for biology and new energy technologies

The important thing in science is not so much to obtain new facts as to discover new ways of thinking about them.

Sir William Henry Bragg
Nobel Prize For Physics, 1915

There is a growing body of scientific evidence that light-initiated reactions in the molecular components of photosynthetic organisms can display quantum-coherent behaviour in their dynamics at ambient conditions. In the simplest picture, quantum-coherent contributions to the dynamics of electronic excitations imply that excitation transfer can be accompanied by superpositions of quantum states sustaining well-defined phase relationships for times long enough to affect transport at the molecular scale.

The experimental observation of coherence oscillations during energy transfer dynamics has ignited interest (and controversy) in the possible biological function of such quantum-coherent behaviour, and also in the way it could be exploited for efficient and robust man-made devices powered by light.

Understanding the importance of quantum-coherent dynamics for biological function boils down to assessing whether the presence of such dynamical phenomena is not just sufficient but necessary for the control and regulation of energy of transport in molecular systems that adapt their function to changing environmental conditions. Independently of whether biological systems do actively use such coherent dynamics, it is of great importance to propose ways in which these quantum phenomena can be harnessed in novel technologies that provide efficient and sustainable concentration, distribution and conversion of solar power at the nanometre scale [1,2]. These two fundamental scientific challenges can therefore only be successfully addressed under the scrutiny of a multi-disciplinary perspective open to new and possibly counterintuitive ideas, while keeping a healthy scepticism that encourages clarity and simplicity of scientific explanations.

In recent years, it has been recognized that a broader understanding of the advantages of quantum-coherent energy transfer can be gained by bridging the gap between two scientific views: the physical chemistry perspective of quantum dynamics, which has a marked emphasis on theoretical descriptions capable of

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predicting spectroscopic signals, and the conceptual perspective of contemporary quantum physics, where quantum coherence can be quantified, and is a resource for emerging quantum technologies.

Along these lines, the principal aim of the Royal Society’s Theo Murphy International Scientific Meeting on quantum-coherent energy transfer and its implications for biology and new energy technologies was to gather an interdisciplinary community to discuss current developments and future directions of this fascinating scientific field. In particular, the meeting was set to discuss the following pivotal questions:

Q1. What are the necessary experimental techniques in order to probe coherent quantum dynamics in natural complexes?
Q2. What are the physical mechanisms supporting such quantum-coherent dynamics?
Q3. What unique roles can these dynamical quantum phenomena play for efficient energy transfer in molecular-scale systems?
Q4. Can coherent photo-excitation transport improve the performance of organic-polymer-based energy technologies?

The meeting gathered leading researchers in the field of photosynthesis and energy transfer.1 Graham R. Fleming and Richard Friend set the scene for the 2 day discussion by presenting, respectively, the perspectives of coherence in natural light-harvesting and fundamental issues of energy conversion in molecular semiconductors. This was followed by presentations of traditional and contemporary theoretical frameworks eloquently discussed by Bob Silbey and Martin Plenio. The afternoon session on experimental methodologies for control and spectroscopy of energy transfer was given by Valeria Kleiman and Shaul Mukamel, and complemented by a general overview from a biological perspective provided by Richard Cogdell. The second morning of discussion was enthusiastically opened by Greg Engel, who described experimental evidence for quantum transport in photosynthetic systems, followed by Yositaka Tanimura, who presented non-perturbative theories of quantum dynamics. A critical perspective on the importance of coherence and entanglement in photosynthesis was given by Marcus Tiersh, and the morning session ended with a discussion in the style of Seth Lloyd about congruence of energy scales and complexity. During the final afternoon session, chaired by Jenny Nelson, Laura Herz and Dohgo Kim shifted the focus towards energy transfer in polymers and synthetic molecular assemblies. The meeting was concluded by Gregory D. Scholes, who lively presented a much needed summary on the lessons we can learn from nature about solar light-harvesting.

The thought-provoking speaker contributions were complemented both by inspiring poster presentations from younger scientists and by the challenging and important issues raised by about 100 enthusiastic participants, representing a variety of scientific areas including biology, physics, chemistry, quantum information and engineering. A report on the meeting was published in Nature Physics [3].

1Audio-records of all presentations and the overview discussion of the meeting can be found at http://royalsociety.org/events/2011/quantum-coherent-energy/.

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1. Discussions contributed to this issue

The collection of papers presented in this issue provides viewpoints on the pivotal questions discussed during the meeting. In what follows, we briefly summarize the scientific contributions of the articles and invite the reader to examine them thoroughly.

(a) Experimental techniques probing quantum dynamics in natural systems

Two-dimensional spectroscopy is the experimental technique used so far to provide evidence of coherent dynamics of excitations in photosynthetic antenna complexes via ensemble measurements. Different details of this methodology are summarized and discussed in the articles by Dawlaty et al. [4] and Hayes & Engel [5], while diagrammatic approaches for the design and simulation of general nonlinear optical responses are illustrated by Biggs et al. [6]. There is, however, an ongoing discussion on the complexity and limits of the interpretation of quantum behaviour in ensemble-averaged measurements. This issue is further explored in the article by Dawlaty et al. [4], in which an application of coherent single-molecule spectroscopy [7] is presented as an alternative.

(b) Theory of mechanisms supporting coherent dynamics at ambient temperatures

Traditionally, the theoretical description of electronic energy transfer relies on the spin-boson model. Basic approximations and physical insights gained by this treatment are revised by Zimanyi & Silbey [8]. However, understanding of the mechanisms preserving coherent dynamics in biomolecular systems requires theories that include correlations between electronic states and the vibrational environment that naturally mediates energy transfer. How the implicit non-Markovian nature of the dynamics under these conditions may be manifested in nonlinear optical measurements is examined in the article by Dijkstra & Tanimura [9], while the defining active role of the ‘noisy’ environment is discussed by Chin et al. [10].

(c) Characterization of quantum features relevant for excitation dynamics

Quantification of coherent contributions to dynamics is fundamental if quantitative relations between transport performance and coherence are to be made [11]. A first attempt has been to quantify electronic coherence through metrics derived in the context of quantum information science [12]. In this Meeting Issue, the article by Smyth et al. [13] compares different measures of exciton delocalization. This subject is critically assessed in the article by Tiersch et al. [14], concluding that further research is needed to identify figures of merit that unambiguously show the advantages of quantum behaviour.

Electronic interactions in closely packed molecular aggregates give rise naturally to collective quantum phenomena that are actively used to enhance energy transfer rates between different complexes. In the article by Abasto et al. [15], symmetry-enhanced transfer is discussed under the effects of disordered and fluctuating phonon environments.
(d) Coherent photo-excitation transport in organic-polymer-based technologies

Useful energy production by natural and man-made systems relies not only on concentrating and distributing solar light energy but also on generating charge carriers for chemical energy storage. Cogdell et al. [16] give a brief overview of how the fundamental reactions in photosynthesis can be split into steps that can possibly be emulated by synthetic versions. In contrast to natural systems, however, current organic-polymer technologies are based on large aggregates of materials where separating the domains of excitation and charge transfer is not simple. The articles by Schmid et al. [17] and Yang & Kim [18] discuss examples of synthetic candidates for excitation transport and their photo-physical properties. Coherence in these systems is an issue that requires further investigation [19].

2. Open problems

We would like to conclude this Preface by pointing out some open problems that were highlighted during the overview discussion at the meeting.

Refined experimental techniques able to tease out the quantum character of energy transport dynamics in natural and synthetic molecular systems are required. In particular, there is a need for experimental approaches capable of probing the detailed structure of exciton–phonon interactions as well as the quantum mechanical features of the vibrational environment itself.

Theoretical proposals putting forward alternative thinking on energy transfer are also important. In particular, theories that can clearly pinpoint the advantages that quantum-coherent phenomena can bring to performance, control and robustness of energy transfer and conversion are needed. Furthermore, theoretical bounds on quantum-enhanced performance of energy transport can be useful for defining how quantum-coherent effects can be controlled in natural systems and enhanced in their synthetic counterparts.

The overall management of solar energy conversion in photosynthetic systems and man-made assemblies involves more than light harvesting. Charge separation, photoprotection and adaptability to changing environmental conditions are fundamental features of robust and efficient energy conversion devices. Can we harness quantum-coherent contributions to dynamics to improve these functions?

Finally, it would be interesting to carry out meaningful comparisons of the energy conversion efficiencies of natural photosynthesis and present photovoltaic technologies [20] assuming that coherent dynamics may be transiently present at different steps of the energy conversion process.

We hope that the collection of articles in this Meeting Issue inspires further research into the unforeseen and far-reaching implications of quantum phenomena displayed by the molecular components of living organisms.
References